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Title: INITIAL UC Davis Reactor Analysis Context

Author(s): Mendoza, Paul Michael

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INITIAL UC Davis Reactor Analysis Context

MNRC Safeguards, Core Cooling, and Calorimetric laboratory - 3.0 hours.

Contents

1	Introduction Activation Constraints/Conditions				
2					
3 Analysis Background					
	3.1	Time Since Removal from Reactor	2		
	3.2	Production of ¹³⁷ Cs	4		
	3.3	Fuel Burn-up (Direct Calculation)	5		
	3.4	Fuel Burn-up (Typical Fission Product Ratio Calculation)	6		
	3.5	Fuel Burn-up (Atypical Fission Product Ratio Calculation)	8		
	3.6	TRU concentrations in the fuel	8		
	3.7	Decay Heat/Dose Calculations	8		

1 Introduction

The INITIAL reactor experiment pitched to UC Davis and agreed upon by the professor teaching the course is a natural uranium neutron activation experiment in the McClellan Nuclear Research Center (MNRC) reactor. Both non-cadmium covered (Ex:1 – bare) and cadmium covered (Ex:2 – Cd) experiments will be performed and analysis will ensure on gamma spectra collected from both experiments. The following provides necessary information for completing the experiment analysis. This information includes definitions, assumptions, and plots tailored to the specifics of this experiment. The content is for the students to be completing the laboratory for reference.

2 Activation Constraints/Conditions

Material	- The material to be irradiated is natural uranium dioxide			
	- Chemical Form - UO_2			
	- Density - 10.97 g/cc			
	- Enrichment 0.72 at.% $^{235}\mathrm{U}$			
	- Mass - 10 grams			
Irradiation Time	- Unless otherwise specified:			
	1. Bare Experiment: 4.6 hours			
G 1.	2. Cd experiment: 24 hours			
Cooling Time	- Unless otherwise specified:			
Time	1. Bare Experiment: $\sim 30 \text{ days}$			
	2. Cd experiment: $\sim 30 \text{ days}$			
Neutron Scalar Flux	 Unless otherwise specified: 1. Bare Experiment: ~ 8.75×10¹¹ n/(cm²·s) 2. Cd experiment: ~ 5.75×10¹¹ n/(cm²·s) 			

3 Analysis Background

3.1 Time Since Removal from Reactor

To verify the operators declaration of time since removal from the reactor, Figures 1 and 2 are provided. These figures show the absolute fraction of activity due to different isotopes in the system for both experiments. When determining relative activities, gamma peaks shown in Table 1 should be used. These peaks are chosen so that interferences are minimalⁱ

If the professor is nice enough to declare a cooling time for the fuel, then Figures 1 and 2 are used for verification. This is done by determining relative activitiesⁱⁱ for isotopes that can be seen in the gamma spectrum.

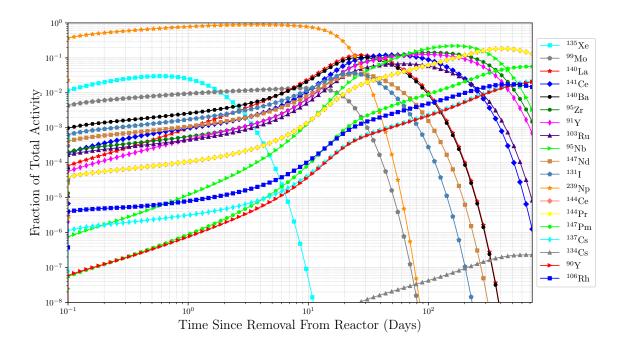


Figure 1: Activity fractions as a function of time since removal from reactor as determined with ORIGEN (Bare Experiment).

If a cooling time is not declared, then it is suggested to first determine the relative activitiesⁱⁱⁱ of ²³⁹Np and another isotope that is increasing (ex. ¹⁴⁴Pr, ¹³⁷Cs, or ¹⁰⁶Rh) and determine whether cooling time is greater than or less than their Cross Over Time (COT, time when activities are equal, ex. COT for ²³⁹Np and ¹⁴⁴Pr is near 30 days (Bare Experiment) and 40 days (Cd Experiment))^{iv}. Once the region of decay time (either greater or less than COT) is established, then more isotopes can be brought into the mix to narrow down exactly when the material was removed from the reactor.

ⁱAn example interference is ¹⁴⁷Pm's largest yield gamma peak at 121 keV. This peak is not included in Table 1 because it over laps with ¹⁴⁷Nd 120 keV peak, and ¹⁴⁰Ba 119 keV peak. ¹⁴⁴Ce is not included in this list because a number of its peaks interferes with with higher activity isotopes (Note: Made interference calculations at 30 days decay (wrote an awesome code to do this)).

ii Described in the appendix

 $^{^{\}mathrm{iii}}\mathrm{Again},$ in the appendix

^{iv}Hint: If $A_{239_{Np}} > A_{144_{Pr}}$ then T < COT. If you can't see ²³⁹Np, this is a good indicator that T > COT.

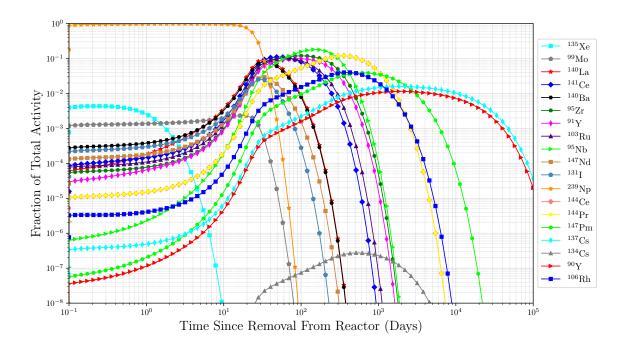


Figure 2: Activity fractions as a function of time since removal from reactor as determined with ORIGEN (Cd Experiment).

Table 1: Gamma energy peaks to use for time since removal from reactor verification.

Isotope	$\lambda~(\mathrm{s}^{\text{-}1})$	$\mathrm{Energy}[\mathrm{keV}](\mathrm{Yield}[\%])$
140 La	4.78×10^{-06}	1596(95.4), 487(45.5), 816(23.3)
$^{143}\mathrm{Pr}$	$5.91{ imes}10^{-07}$	$742(1.2{ imes}10^{-06})$
$^{141}\mathrm{Ce}$	$2.47{\times}10^{\text{-}07}$	145(48.2)
$^{140}\mathrm{Ba}$	$6.29{\times}10^{\text{-}07}$	537(24.4), 30(14.1), 163(6.2)
$^{95}{ m Zr}$	$1.25{\times}10^{\text{-}07}$	757(54.0), 724(44.2), 236(0.3)
$^{91}\mathrm{Y}$	$1.37{ imes}10^{-07}$	1205(0.3)
$^{103}\mathrm{Ru}$	$2.04{ imes}10^{-07}$	497(90.9), 610(5.8), 444(3.3)
$^{95}{ m Nb}$	$2.29{\times}10^{\text{-}07}$	$766(100.0),\ 204(2.8{\times}10^{\text{-}02})$
$^{147}\mathrm{Nd}$	7.31×10^{-07}	91(28.0),531(13.1)
$^{131}\mathrm{I}$	1.00×10^{-06}	364(81.7), 637(7.2)
$^{239}\mathrm{Np}$	$3.40{ imes}10^{-06}$	106(27.2)
$^{144}\mathrm{Pr}$	$6.69{ imes}10^{-04}$	697(1.3), 2186(0.7), 1489(0.3)
$^{132}\mathrm{I}$	$8.39{ imes}10^{-05}$	773(75.6), 955(17.6)
$^{147}\mathrm{Pm}$	$8.38{ imes}10^{-09}$	$76(1.2\times10^{-08})$

3.2 Production of ¹³⁷Cs

Several samples of natural uranium (0.72 at.% ²³⁵U) are to be irradiated in the neutron flux spectra of the MNRC. In the course of irradiation the composition of the fuel will change due to fissioning of fuel into fission products (FP), transmutation of nuclides from neutron absorption to create TRansUranics (TRU – elements with more protons than uranium), and radioactive decay (among other things). The concentration as a function of time of each component in the fuel (U, TRU, FP) can be described with a large system of differential equations. For example, the concentration of ²³⁵U in the system can be described under the 1-group **assumption** with Equation 1.

$$\frac{\delta N_{235}(t)}{\delta t} = -(\lambda_{235} + \phi \sigma_{235}) N_{235}(t) \tag{1}$$

where:

 N_{235} = the number of 235 U atoms in the presence of the scalar flux.

 $\lambda_{235}=$ the radiological half-life of $^{235}\mathrm{U}$

 ϕ = the total scalar flux and

 $\sigma_{235}=$ the average^{vi} total transmutative cross section for ²³⁵U (absorption, fission, (n,2n), etc.)

If we assume ϕ and σ are constant as a function of time, then the solution to Equation 1 is a simple exponential.

$$N_{235}(t) = N_{0,235}e^{-(\lambda_{235} + \phi\sigma_{235})t}$$
(2)

As a matter of fact, the solution to every isotope in the system is an exponential of one form or another. In the case of Equation 2, if we **assume** short irradiation times, then $N_{235}(t) \approx N_{0,235}$. The assumption that the number of ²³⁵U atoms in the system is constant will help with the derivation for the concentration of one of the fission products.

In the case where only $^{235}{\rm U}$ is undergoing fission, the differential equation for the concentration of $^{137}{\rm Cs}$ could be written as shown in Equation 3.

$$\frac{\delta N_{137}(t)}{\delta t} = -(\lambda_{137} + \sigma_{137}\phi)N_{137}(t) + \gamma_{137}N_{0,235}\phi\sigma_{f,235}$$
(3)

where:

 γ_{137} = the cumulative fission yield for ¹³⁷Cs $\left[\frac{\text{\# of }^{137}\text{Cs atoms}}{\text{fission of }^{235}\text{U}}\right]^{\text{vii}} \approx 0.0627$

 σ_{137} = the average total transmutative cross section for $^{137}\mathrm{Cs} \approx 0$

 $\sigma_{f,235}$ = the average fission cross section for ²³⁵U

Note: **Assuming** for γ_{137} that all fission isotopes in the 137 mass bin immediately arrive at ¹³⁷Cs without passing through any of the precursors (precursor examples include: ¹³⁷Xe, ¹³⁷I, ¹³⁷Te).

^vAlso **assuming** homogeneous flux and material. Additionally that the sample being irradiated does not perturb (or change) the flux spectrum.

vi In this context "average" refers to the flux averaged (over energy) cross section $\sigma = \int \sigma(E)\phi(E)dE/\int \phi(E)dE$. vii Note: Will **assume** this is constant for all fissioning isotopes and energies of neutrons causing fission.

Given the assumptions above the solution for the number of 137 Cs atoms as a function of time is given in Equation 4^{viii} .

$$N_{137}(t) = \gamma_{137} N_{235} \phi \sigma_{f,235} t \tag{4}$$

3.3 Fuel Burn-up (Direct Calculation)

Fuel burn-up can be defined in several ways. One way is an operational parameter for the amount of energy produced per unit mass of the fissionable material. For a constant power, burnup is shown in Equation 5.

$$BU = \frac{\text{Power [MW]} \cdot \text{days}}{\text{mass [tHM]}}$$
 (5)

where:

Power = thermal power released into the working fluid

days = number of days operated at Power

mass = the initial mass of heavy metal in the irradiated fuel in metric tons

In order to estimate burnup with fission cross sections and number densitites of fissionable material, Equation 6 can be used (with units and values showed for clarity and for future calculations).

$$BU\left[\frac{\text{MWd}}{\text{tHM}}\right] = \frac{C\left[\frac{\text{MWd}}{\text{MeV}}\right] \cdot \phi_{tot}\left[\frac{n}{cm^2 \cdot s}\right] \cdot T[s]}{m_{HM}[\text{tons}]} \cdot \sum_{i} N_i(\sigma_{f,i}E_f + \sigma_{\gamma,i}E_{\gamma}) \left[\frac{cm^2 \cdot \text{MeV}}{n}\right]$$
(6)

where:

E = Energy released per fission (f) or capture (γ) reaction

1. Fission: Assuming 200 MeV/fission for all isotopes

2. Capture: $^{235}U - 6.5 \text{ MeV}$, $^{238}U + 4.5 \text{ MeV}$

 σ_{γ} = capture single-group neutron cross section

1. Bare experiment: $^{235}U - 30 \text{ b}$, $^{238}U - 5.6 \text{ b}$

2. Cd experiment: $^{235}U - 4.3 \text{ b}, ^{238}U - 7.4 \text{ b}$

 σ_f = fission single-group neutron cross section

1. Bare experiment: $^{235}U - 162 \text{ b}$, $^{238}U - 0.076 \text{ b}$

2. Cd experiment: $^{235}U - 9.75$ b, $^{238}U - 0.12$ b

 ϕ_{tot} = total scalar neutron flux

1. Bare experiment: $\phi_{therm}/f_{therm} = 3 \times 10^{11}/0.34$ $f_{therm} = 0.34$

2. Cd experiment: $\phi_{tot} \cdot (1 - f_{therm}) = 5.75 \times 10^{11} \text{ n/(cm}^2 \cdot \text{s)}$

 N_i = Atoms: 235 U - 1.61×10²⁰, 238 U - 2.2×10²² (10 grams of Nat. UO₂)

viiiWhy am I ignoring the $(\lambda_{137} + \sigma_{137}\phi)N_{137}(t)$ term on the RHS of Equation 3, and what two assumptions are included in this ignoration?

C = Energy conversion factor $1.9 \times 10^{-24} \left[\frac{\text{MWd}}{\text{MeV}} \right]$

 m_{HM} = the metric tons of heavy metal in the system (8.8×10⁻⁶ tons for 10 grams of Nat. UO₂)

i = index for fissionable isotopes in the system (Hint: i will equal 235 U and 238 U - we can ignore 239 Pu)

T = the irradiation time.

Note that many of the assumptions that applied to Equation 4 also apply to Equation 6. Also the connection between Equation 5 and Equation 6 can be made by understanding that the term, $\phi N \sigma E$, has units of MeV released per second.

We can determine burnup from our measurements without prior knowledge of the neutron flux spectrum. Equation 7 shows how this can be done^{ix}.

$$BU = \frac{N_{137}}{N_{0,HM}} \cdot \frac{N_A E_f C}{\gamma_{137}} \left[\frac{MWd}{mols} \right] \cdot \frac{1}{M_0^{HM}} \left[\frac{mols}{tHM} \right]$$
 (7)

where:

 $N_A = \text{Avogadro's constant}$

 $M_0^{HM} = \text{the initial heavy metal molar mass } (\sim 2.38 \times 10^{-4} \text{ tHM/mol}),$

and all other terms have been previously defined. It should be noted that energy release from radiative capture is not included in Equation 7, where it is included in Equation 6. The effect this will have on burnup estimates is left to the student for explanation.

3.4 Fuel Burn-up (Typical Fission Product Ratio Calculation)

With large fuel assemblies, the absolute activity of $^{137}\mathrm{Cs}$ is difficult to determine due to self-shielding. Another method to determine the burnup of a fuel assembly is to use a ratio of activities, so that absolute amounts of a particular isotope is not necessary. Two common isotope ratios are $^{134}\mathrm{Cs}/^{137}\mathrm{Cs}$ and $^{154}\mathrm{Eu}/^{137}\mathrm{Cs}^{[1, \mathrm{Ch.~18-Sec.~3.5}]}$. These ratios are used because the relationship is linear as a function of burnup.

Figure 3 shows a plot of these two isotopes as a function of burnup for both our experiments and has been calculated using MCNP generated neutron flux spectra coupled with SCALE's ORIGEN module. It should be noted that the logarithmic scale on the y-axis makes the relationship look non-linear and it is also important to note that the slopes for the two different experiments are different. Differences in slope are due to differences in flux spectra (Bare \sim thermal, Cd \sim fast) leading to the use of different fission product yields in the calculation as well as different reactions being favorable.

A final point of note for Figure 3. There are two main production schemes for 134 Cs and 154 Eu. Direct fission product yield to the isotope (ex. Fission \rightarrow 134 Cs) and neutron capture on a lighter nuclei (ex. Fission \rightarrow 133 Cs \rightarrow 134 Cs). In power reactors, the second scheme reflects the majority of 134 Cs and 154 Eu production, whereas in our system, the majority comes from the first. The reason for this is because the total scalar neutron flux is much less in a research reactor (noting also that the second scheme is proportional to the total scalar neutron flux squared).

To determine the burnup with these plots one needs to fit a line to each curve with the origin as an intercept, and determine the appropriate activity ratio.

^{ix}Can you derive Equation 7 from Equations 6 and 4

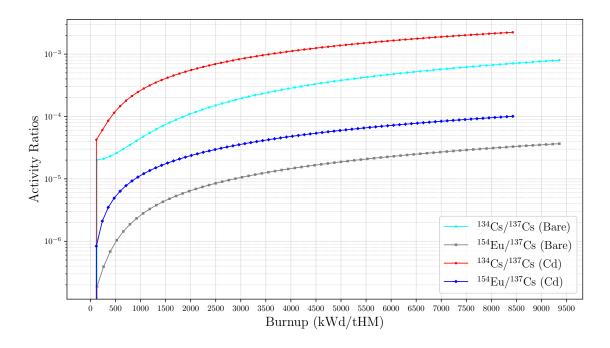


Figure 3: $^{134}\text{Cs}/^{137}\text{Cs}$ and $^{154}\text{Eu}/^{137}\text{Cs}$ activity ratios as a function of burnup as determined with ORIGEN.

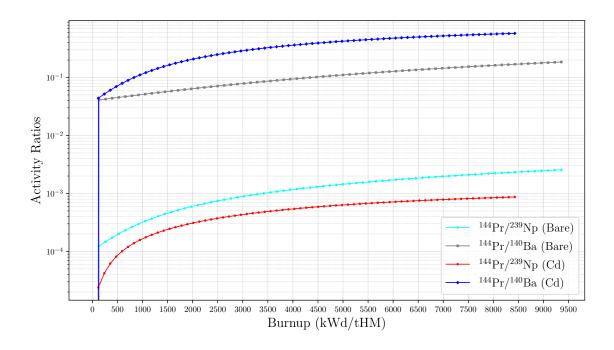


Figure 4: Atypical activity ratios as a function of burnup as determined with ORIGEN.

3.5 Fuel Burn-up (Atypical Fission Product Ratio Calculation)

In our situation, our irradiated UO₂ might not have radioactively cooled long enough for the ¹³⁴Cs, ¹⁵⁴Eu, or ¹³⁷Cs to be distinguishable from all the other high activity fission products. Some ratio plots are provided in Figure 4 for some of the shorter lived isotope ratios as a function of burnup. Please note: <u>Proper</u>^x decay correction to the end of irradiation time is necessary for Figure 3 or Figure 4 to be beneficial^{xi}.

3.6 TRU concentrations in the fuel

Figure 5 shows how ²³⁸U is transmuted via neutron absorption (σ_{γ}) and radioactive decay (β^{-}) during irradiation to produce different plutonium isotopes. It could imagined how heavier isotopes and elements could be created in the system.

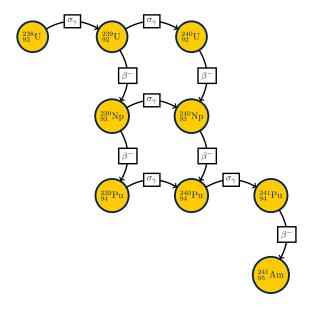


Figure 5: A transmutation network for ²³⁸U.

Describing such a system with a series of differential equations is possible, and analytical solutions exist for each isotope, but this can get involved. It should be known though, that as neutron fluence increases, the mass of the TRansUranics (TRU) will increase as uranium is converted to the heavier isotopes. These heavy isotopes are unstable and will mostly undergo alpha decay with small probabilities for spontaneous fission.

3.7 Decay Heat/Dose Calculations

Residual heat in irradiated fuel is due to radioactive decay. As described previously, fission products^{xii} and TRansUranics^{xiii} are produced during the course of irradiation. These heavy and "light" radioactive species emit different radiation. The longer lived heavier elements mostly decay by α (⁴He) emission or through spontaneous fission. Both processes also emit photons. The fission products mostly decay by beta (a fast electron) emission. This process also emits

 $^{^{}x}$ Don't decay correct the 144 Pr (notice its really short half-life), but rather the 144 Ce that feeds the 144 Pr.

 $^{^{\}rm xi}$ Hint: You might get better answers using the 239 Np normalization if you can see its peak. If you can't see the 106 keV peak for 239 Np then use the early portion (<1000 kWd/tHM) of the 140 Ba normalization lines.

 $^{^{\}text{xii}}$ Example of 137 Cs used in Section 3.2

 $^{^{\}rm xiii}{\rm Example}$ of Pu used in Section 3.6

photons. For a visual representative, the Bare experiment was simulated in ORIGEN, and the fraction of decay heat associated with these radiations has been plotted as a function and is shown in Figure 6.

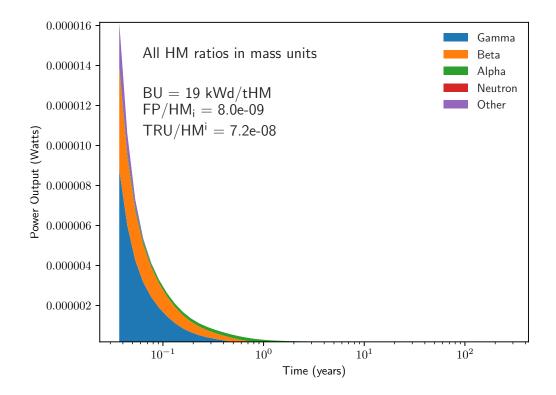


Figure 6: Decay heat plot for bare experiment.

Notice how Figure 6 shows that for our experiment, most of the decay heat radiation comes from the gamma emissions. The power output is also very small because of the small mass of fuel irradiated. It should be noted that all this decay heat will **not** deposit inside the fuel because gamma radiation has a good chance of escaping the material.

So that we do not get the wrong idea, Figure 7 is provided to show what the decay heat signature would look like if our bare experiment irradiation continued until the burnup was comparable to power reactor fuel burnup. It should be noted the differences in overall watt output, burnup, fission product (FP) mass and TRU mass in either scenario, as these might be helpful for answering laboratory questions.

To estimate the dose (or heating) from each of these species for a **short irradiation experiment**, it would be helpful to assume that alpha and neutron emissions are negligible. Further, once a time of decay has been estimated for both experiments, Figures 1 and 2 would be helpful for determining activities of any species that could not be determined with a gamma spectrum (one would need to determine the absolute activity of a single isotope and use that isotope, along with either Figure 1 or 2 – depending on experiment – to determine the activity of the other species in the fuel). Once the activity contributors have been determined, a useful tool for determining how much heat a particular isotope is emitting is an Evaluated Nuclear Data File (ENDF). Specifically, MF 8, MT 457. If those numbers do not make sense, then follow this link: http://www.nndc.bnl.gov/exfor/endf00.jsp and use Figures 8 – 10 in the appendix

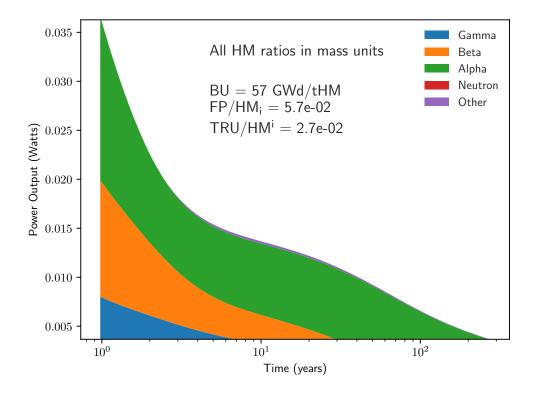


Figure 7: Decay heat plot for bare experiment.

for help in navigation^{xiv}.

The **total** energy emission could be determined with each isotope using this information. Equation 8 shows how this would be done for an individual isotope. For gamma energies, the energies associated with gammas, x-rays, and 511 annihilations would need to be substituted for the $Q-E_{\rm neutrino}$ term.

$$Heat = A \cdot (Q - E_{neutrino}) \tag{8}$$

References

 DT Reilly, N Ensslin, HA Smith Jr, and S Kreiner. Passive nondestructive assay of nuclear materials (panda) manual. Technical report, NUREG/CR-5550 (US Government Printing Office, Washington, DC, 1991). Los Alamos National Laboratory document LA-UR-90-732, 1991.

xiv The example in the figures is ⁶⁰Co, please substitute for your own isotopes of interest

Appendix

Relative Activities

Absolute activities will be needed for the decay heat calculation. For the time since removal calculation, a relative activity could be used. For example, using the 1205 keV peak for 90 Y, and the 106 keV peak for 239 Np. The relative activity for 239 Np, among these two isotopes could be determined with Equation 9.

$$R_{A,239} = \frac{A_{239}}{A_{239} + A_{90}} \tag{9}$$

Where a single activity is determined with Equation 10. Note that if the geometric efficiency is not known, it does not matter because its value cancels out when plugged into Equation 9, only an energy calibration is needed.

$$A_{239} = \frac{CPS_{1205}}{BR_{1205}\epsilon_{Energy=1205}\epsilon_{geo}}$$
 (10)

In order to use this information to determine the time since removal from reactor, find the fraction of activity of these two isotopes at your suspected time (use Figure 1 or 2), and add use those fractions, f_{239} and f_{90} , to determine another relative activity, shown in Equation 11. If the relative activities determined from Figure 1 or 2 match with the experimentally determined value, then the suspected time since removal from reactor is correct. If not, then another time should be subject to the same methodology. The shape of the graphs might be able to inform what the next guess of time since removal from reactor should be. This method can be expanded to include as many isotopes as measurable.

$$R_{A,239} = \frac{f_{239}}{f_{239} + f_{90}} \tag{11}$$

As a potentially helpful aside for students, if an absolute activity for a single species were known along with the time since removal from reactor. Then the activity of another species in the system could be estimated with Equation 12.

$$A_2 = A_1 \frac{f_2}{f_1} \tag{12}$$

Brookhaven site ENDF Navigation

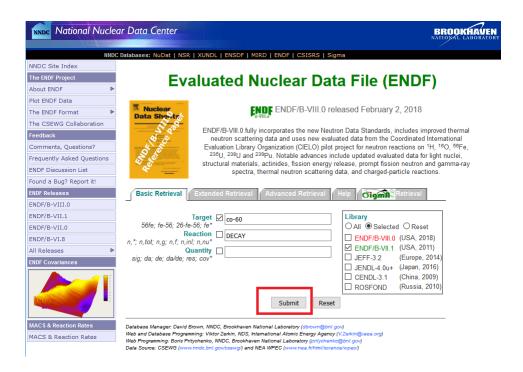


Figure 8: Link Provided leads to this page.



Figure 9: Clicking on "Submit" as shown in Figure 8 leads here.

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Figure 10: Clicking on "Info" as shown in Figure 9 leads here, where the data is provided.